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STRUCTURES OF URANIUM EXTRUDED HIGH IN THE ALPHA PHASE

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Conditions were explored for extruding uranium at temperatures high enough in the alpha phase for at least the core of the extruded rod to be transformed into the beta phase as it leaves the die. This uranium then is similar in grain size and texture to uranium conventionally subjected to a separate post-extrusion beta treatment.

Les conditions de l'extrusion de l'uranium aux températures assez élevées du domaine α ont été étudiées de telle façon qu'au moins le cœur du barreau extrudé soit transformé en phase beta quand le barreau sort de la filière. Cet uranium est alors semblable du point de vue de sa grosseur de grain et de sa texture à l'uranium soumis de manière classique à un traitement distinct en phase β postérieur à l'extrusion.

Es wurden die Bedingungen untersucht, unter denen man Uran in einer so hohen Temperatur der α-Phase strangpressen kann, dass mindestens der innere Teil des Presslings sich in die β-Phase umwandelt wenn er die Strangpressform verlässt. So bearbeitetes Uran gleicht in der Korngrösse und Textur dem konventionellen Uran, welches nach dem Strangpressen einer besonderen β-Behandlung unterzogen wurde.

1. Introduction

For dimensional stability under irradiation, the grains of metallic uranium should be both randomly oriented and fine: preferred orientation causes anisotropic growth; coarse grains lead to rough surfaces (bumping, wrinkling, dimpling). Fine grains and random orientation are somewhat incompatible objectives since the means of improving one property is likely to impair the other: working to reduce grain size also increases the preferred orientation; heat treatment to randomize the structure by transforming the uranium into the beta phase coarsens the grains. In practice, primary consideration is given to achievement of a random structure through beta treatment of material worked in the alpha phase; reduction of grain size is sought by controlling the rate of cooling and by alloying. Although rapid cooling is desirable for grain refinement, excessively rapid cooling can introduce preferred orientation.

The program reported here was undertaken to explore the possibility of obtaining novel structures, possibly combining random structure and fine grains, by extruding uranium at temperatures high in the alpha phase. The texture (and hence the irradiation growth) of uranium is known to be strongly dependent on the temperature of alpha working [1]). In the

† The rise in the transus is of course associated with the volume increase in the phase change. Calculation of the magnitude of the temperature increase from the Clausius-Clapeyron equation ($\Delta P/\Delta T = \Delta H/T\Delta V$) gives $\Delta P/\Delta T = 3460$ psi/°C. Thus, under a stress of 100 000 psi, the transformation to beta uranium requires a temperature higher by about 30° C. The heat generated in extrusion is certainly capable of effecting such a temperature increase in the uranium. The exact temperature increase depends on dissipation of heat to the tooling. Higher temperatures are expected along the axis of the billet and the extruded rod.

In 1944 Creutz and Gurinsky [3]) cited the possibilities of the increase of the uranium temperature by extrusion work and the raising of the alpha-beta transus by the extrusion pressure. Their Clausius-Clapeyron calculation gave a $\Delta P/\Delta T$ of about 4000 psi/°C. The value of 3460 given above is in excellent agreement because of cancellation of the effects of downward revision of both ΔH and ΔV from the values available in 1944.

light of earlier observations in this laboratory on extruded uranium, it appeared that a substantial portion of the uranium could reach temperatures above the alpha-beta transus, thus undergoing beta treatment immediately subsequent to the extrusion. Such transformation as a result of heating during alpha working has been reported during rolling at 630° C [2]). The transformation is believed to be delayed until after the metal leaves the die and is no longer subject to the extrusion pressure which has raised the alpha-beta transus †. The consequent beta treatment can affect all of the extruded metal, which then acquires the typical structure of beta-treated uranium, with a random orientation but relatively coarse grains. It was also found that an interesting duplex structure could be obtained: the bulk of the rod acquired the beta-treated structure; it was surrounded by a rim of fine-grained, textured metal that apparently had not been heated sufficiently to undergo transformation into the beta phase. To extend the application of the method of achieving a suitable combination of minimum texture and minimum grain size, alloying with silicon (0.16 wt %) was also tried.

2. Materials and procedures

2.1. Materials

Two billet compositions were used in the extrusion experiments: unalloyed ingot uranium and a uranium-0.16 wt % silicon alloy. Most of the extrusions were made with the unalloyed uranium. Both billets were triple-beta-treated with a water quench to eliminate effects of prior working history.

2.2. Extrusion

The billets were enclosed in 16-gage (1.63 mm) copper cans, heated to the extrusion temperature and extruded from a 2.800″ (71 mm) dia. container through a die having a diameter in the range of 0.5″ to 0.9″ (12 to 23 mm). Details of the extrusion conditions are presented in table 1. Heating time for the billets was 3 h and temperatures were held to within ± 5° C.

2.3. Post-extrusion cooling

The copper-clad uranium rods went from the press into a steel catch tube, where they cooled in still air or in a water quench. Two types of water-quenching devices were used: 1. In one, the rods were exposed to water in the catch tube when they penetrated a rubber diaphragm at 10″ (254 mm) from the die. 2. In the other, the rods were sprayed with water as they left the die, starting at one inch from the die. The diaphragm device was used in the initial extrusions; the spray device, which gives a more drastic quench, was installed to reduce the grain size of the extruded rods.

2.4. Methods of evaluation

Short sections were cut from the extruded rods to obtain specimens for metallographic examination and X-ray diffraction studies of the uranium. The results listed in table 1 are for specimens from the mid-length of the rods; in a few rods, specimens were cut from the front and rear of the rod for comparison with the specimens from the middle.

2.4.1. *Metallographic examination*

The grain size and the general appearance of transverse sections from the rods were viewed at two magnifications. A magnification greater than 50 × (micro) was used on specimens with grains smaller than 100 μm; a low magnification of 3 × (macro) was used on specimens with grains larger than 100 μm.

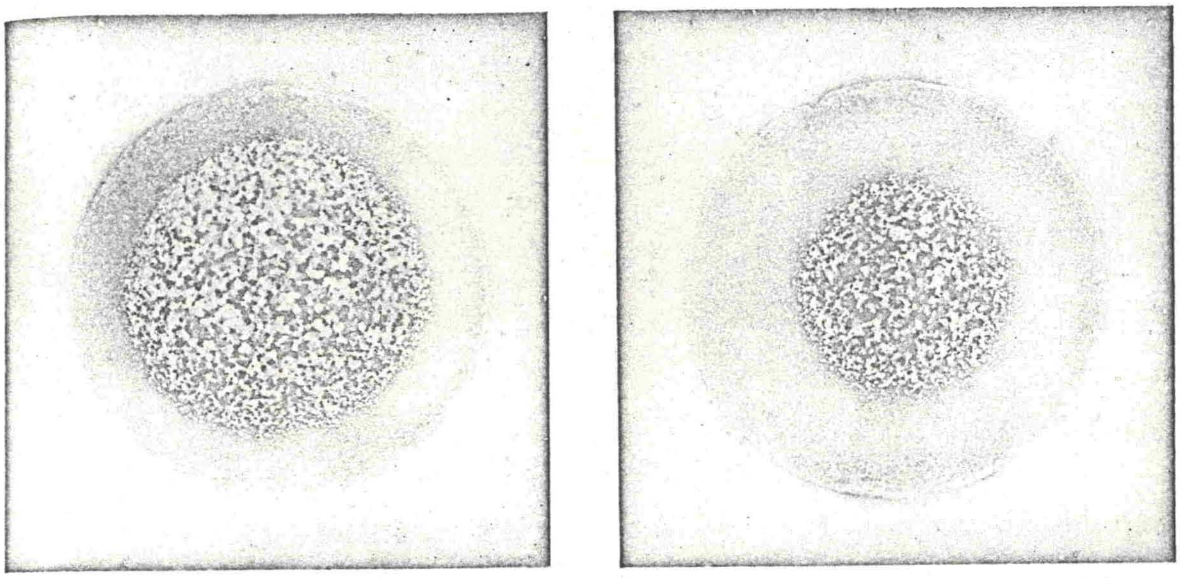
2.4.2. *Crystallographic orientation*

Crystallographic orientation was determined by an X-ray diffraction technique in which the intensity of significant poles was measured relative to the intensity of the poles of a random sample. The crystallographic orientation is expressed in table 1 in terms of longitudinal growth index values (GI_L) [4]). The GI_L value is related to the expected positive or negative anisotropic growth in the longitudinal direction in nuclear irradiation; one unit of GI_L is equivalent to one percent dimensional change per 1000 ppm burn-up for ordinary beta-

TABLE 1
Conditions and results for uranium extruded high in the alpha phase

No.	Composition [1]	Billet temp. (°C)	Reduction ratio	Ram speed (in/min)	Strain rate (sec^{-1})	Container temp. (°C)	Extrusion constant K (tsi)	Quench [2] distance (in)	Grain size (μm)		$GI_{long.}$				
									Core	Rim	1	2	3	4	avg.
Group 1. *Structures that appear to have transformed from the beta phase (beta structure)*															
1	U	650	40	110	73	500	18	1	200	100	+2.3	+2.5			+2.4
2	U	650	40	110	73	260	19	10	500	500	+0.3	−0.2	+0.2	−0.2	0
3	U	650	25	110	46	500	20	1	750	750	+1.1	−1.8			−0.3
4	A	650	25	110	46	500	24	1	50	50	−0.1				
5	A	650	25	110	46	500	24	1	50	50					
6	U	650	40	55	37	500	25	1	400	400	+1.8	+1.6			+1.7
7	U	650	25	55	23	500	20	1	250	250	+1.8	+0.9			+1.4
8	U	650	40	20	13	260	17	10	1000	500	+4.5	+3.5			+4.0 [3]
9	U	593	40	110	73	500	19	1	1000	1000	−1.3	−1.9			−1.6 [4]
10	U	593	40	110	73	500	18	10	500	500	−0.3	−1.7			−1.0
11	U	650	40	130	86	500	19	air cool	> 1000	> 1000					
12	U	650	40	110	73	500	21	air cool	> 1000	> 1000					
Group 2. *Structures that appear to have been alpha worked and recrystallized (alpha structure)*															
13	U	650	10	20	3.3	260	14	10	20	20	−5.4				−5.4
14	U	593	10	110	18	500	15	10	15	15	−5.2				−5.2
15	U	593	10	20	3.3	500	13	10	20	20	−3.9				−3.9
Group 3. *Structures with "alpha" cores and "beta" rims*															
16	U	650	10	110	18	260	17	10	250	20	+0.4	0			+0.2
17	U	650	10	55	9.2	500	16	1			+0.4				+0.4
	Core								500		+0.3	+0.5			+0.4
	Rim									20	−0.5	−1.4			−1.0
18	U	650	10	55	9.2	500	14	1							
	Core								500		+0.7	−0.1			+0.3
	Rim									20	−3.0				−3.0
19	A	650	10	110	18	500		1							
	Core														
	Rim														
20	U [5]	650	10	55	9.2	250		1							
	Core								500		+0.1				
	Rim									20					
21	U [5]	650	10	55	9.2	250		1							
	Core								500						
	Rim									20	−1.6				
22	U	593	40	20	13	500	15	10	250	25	−3.4				−3.4
Group 4. *Rattlesnaked extrusions*															
23	U	675	40	110	73	500									
24	A	650	40	110	73	500									

(1) U=unalloyed uranium; A=uranium-0.16 wt % silicon alloy.
(2) Distance from die to quenching head.
(3) The large grain size of this rod makes the X-ray texture results unreliable. The relative pole intensities varied widely between runs.
(4) Although the grain size is very large, the relative pole intensities do not vary much between runs. This average may be reliable.



(3 ×) RF-7601 (3 ×) RF-7602

Fig. 1. Photomacrographs of cross-sections from the middle of rods 20 (on left) and 21 (on right). These illustrate the quenching effect of a cool extrusion container on the thickness of the fine-grained rim. Rod 21 was the rear billet in a tandem extrusion and was in contact with the cool extrusion container (250° C) for 3 sec longer than rod 20.

treated uranium. Material with a GI_L of zero is sought since it would be expected to be dimensionally stable in irradiation.

3. Results

Two types of microstructure were observed in the extruded rods, singly or in combination. The "beta structure" is similar in appearance to beta-transformed uranium and is characterized by large (50 to 1000 μm depending on composition), irregularly shaped grains with scalloped boundaries (core in fig. 1). The "alpha structure" is similar in appearance to uranium that has been alpha worked and recrystallized; it is characterized by small (less than 50 μm) equiaxed grains with smooth boundaries (rim in fig. 1).†

The extruded rods can be divided into four groups (table 1) depending on the presence of these structures:

Group 1: The rods in this group have a beta microstructure throughout the cross-section. In three rods (6, 7 and 8) that were drastically quenched the grains on the rim are elongated in the radial direction. These three rods have positive GI_L values. In the other rods the grains are more equiaxed though very irregular. The crystallographic orientations of these rods can be described as "random-like", i.e. there is no strong preferred orientation; such a texture is different from the highly preferred orientation (large negative GI_L) ordinarily produced in uranium extruded in the alpha phase.

Group 2: The rods in this group have an alpha microstructure and a crystallographic orientation (large negative GI_L values) that are typical of uranium extruded in the neighborhood of 550° C.

Group 3 (fig. 1): The rods in this group have a core of beta structure surrounded by a rim of alpha structure. The size, shape and orientation of the central grains are the same as for the rods consisting entirely of beta structure. In contrast, the grains in the rim are the same as those in the rods with alpha structure.

† Additional photomicrographs are reproduced in [5]).

Group 4: The rods in this group have a beta microstructure; however, the rods have circumferential cracks. These cracks presumably occurred during extrusion, when sharp variations in extrusion pressure were noted. The circumferential cracks have the characteristic appearance known as a "rattlesnake" defect in extruded shapes.

4. Discussion

The observed structures and textures can be interpreted as consequences of the extent to which the uranium transforms into the beta phase during or after extrusion. The extrusion work can raise the temperature of the uranium above the normal transformation temperature (663° C). Since the transformation temperature is increased by pressure, the extruding uranium can remain in the alpha phase until it leaves the die land. There the uranium transforms into the beta phase, acquiring a random texture and a grain size determined by the rate of cooling from the beta.

The extent of transformation into the beta depends on the severity of the extrusion conditions: initial billet temperature and strain rate (reduction and ram speed). If these conditions are severe enough for complete transformation to occur *after* extrusion, only beta structure is obtained. If the extrusion conditions are so severe as to permit some transformation even *during* extrusion, rattlesnaking results (Group 4). The alpha structures of Group 2 reflect mild extrusion conditions. The Group 3 structures (beta core with alpha rim) are produced by conditions that facilitate cooling of the billet surface while beta temperatures are attained in the core.

A beta-structure rod fast cooled after it leaves the die tends to acquire the fine grain size imparted by a quench after conventional beta treatment. Rods 11 and 12, which were air-cooled, had grains larger than 1000 μm. The smallest grains were found in rods that had been sprayed with water as they left the die. However, in rods 3 and 9, the grain size was 750 and 1000 μm respectively, in spite of the water spray. This large grain size was probably caused by a delay in turning on the spray device, which was hand-operated on a signal from the press operator.

A drastic quench produces radially columnar grains near the surface (rods 6, 7 and 8). These rods had a texture with a concentration of {010} poles *parallel* to the rod axis, giving a positive GI_L. This texture is unusual for uranium extruded at high temperatures in the alpha phase; under these conditions {010} poles usually are *perpendicular* to the rod axis. Work at another laboratory [6]) has shown that alpha grains formed in a steep thermal gradient grow in a columnar fashion with the {100} poles aligning themselves in the direction of the thermal gradient. The {010} axial texture in the quenched rods may thus be a consequence of the {100} radial texture. The unusual direction of the {010} poles in the quenched rods is thus attributed to the effect of the steep radial temperature gradient during quenching.

The grains of "beta structure" rods can also be refined by alloying as is illustrated in the small grain size (50 μm) of uranium-0.16 wt % silicon nos. 4 and 5 compared to the larger grain size (750 μm) of unalloyed rod no. 3 extruded under the same conditions. The thickness of the fine-grained rim can be controlled by cooling the rim of the billet before extrusion. Two billets (rods 20 and 21) were extruded in tandem in a cool extrusion container (250° C). The rear billet was in contact with the cool extrusion container for 3 sec longer after the extrusion upset than was the front billet. The thickness of the fine-grained rim on the rear rod was approximately twice the thickness of the fine-grained rim on the front rod (fig. 1).

5. Summary and conclusions

Extrusion conditions can be selected so that an extruded uranium rod transforms to the beta phase as it leaves the die and thereby acquires a structure that ordinarily is achieved by a separate post-extrusion beta treatment. This beta structure may be present throughout the rod or it may constitute a core surrounded by a rim of fine-grained material, which has not transformed into the beta phase. This rim thus has a preferred orientation and also very fine grains; the latter may help minimize in-pile bumping of the surface by the coarse core grains. The thickness of the fine-grained rim can be controlled through cooling of the rim of the extrusion billet, e.g. through contact with the extrusion tools.

The beta-treated structure achieved directly by extrusion is presumably suitable for isotropic behavior under irradiation. The tendency for surface bumping can be reduced by the fine-grained rim achievable in the same extrusion operation. Such a rim may be unnecessary if the core is alloyed to obtain a finer grained all-beta structure.

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